Biofiltration vs conventional activated sludge plants: what about priority and emerging pollutants removal?

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ABSTRACT

This paper compares the removal performances of two complete wastewater treatment plants (WWTP) for all priority substances listed in the Water Framework Directive and additional compounds of interest including flame retardants, surfactants, pesticides and personal care products (PCP) (n=104). First, primary treatments such as physico-chemical lamellar settling (PCLS) and primary settling (PS) are compared. Similarly, biofiltration (BF) and conventional activated sludge (CAS) are then examined. Finally, the removal efficiency per unit of nitrogen removed of both WWTPs for micropollutants is discussed, as nitrogenous pollution treatment results in a special design of processes and operational conditions. For primary treatments, hydrophobic pollutants (log K_{ow} > 4) are well removed (> 70%) for both systems despite high variations of removal. PCLS allows an obvious gain of about 20% regarding pollutant removals, as a result of better suspended solids elimination and possible coagulant impact on soluble compounds. For biological treatments, variations of removal are much weaker and the majority of pollutants are comparably removed within both systems. Hydrophobic and volatile compounds are well (> 60%) or very well removed (> 80%) by sorption and volatilization. Some readily biodegradable molecules are better removed by CAS indicating a better biodegradation. A better sorption of pollutants on activated sludge could be also expected considering the differences of characteristics between a biofilm and flocs. Finally, comparison of global processes efficiency using removals of micropollutants load normalized to nitrogen shows that PCLS + BF is as efficient as PS + CAS despite a higher compactness and a shorter hydraulic retention time (HRT). Only some groups of pollutants seem better removed by PS + CAS like alkylphenols, flame retardants or DEHP, thanks to better biodegradation and sorption resulting from HRT and biomass characteristics. For both processes, and out of the 68 molecules found in raw water, only half of them are still detected in the water discharged, most of the time close to their detection limit. However, some of them are detected at higher concentrations (> 1 µg/L and/or > Environmental Quality Standards) what is problematic as they represent a threat for aquatic environment.

KEYWORDS
Priority pollutants; emerging pollutants; biofiltration; conventional activated sludge; physico-chemical lamellar settling; wastewater treatment plant; nitrification

INTRODUCTION

The fate of micropollutants in the environment has become an increasing issue last decades, especially in heavily urbanized areas. To struggle against water contamination, the European Community adopted a control policy strategy materialized by the European Water Framework Directive (WFD, Decision No. 2455/2001/EC) (EC 2001). This regulation requires Member States to achieve a good ecological and chemical status in surface waters by 2015, 2021 or 2027. Concretely, 41 chemicals were defined as priority substances because they represent a significant risk for the aquatic environment, and Environmental Quality Standards (EQS) have been set for 33 of them. In parallel of WFD pollutants, a large number of molecules such as pesticides, pharmaceuticals and personal care products (PPCP), flame retardants, etc. are detected in the environment and also represent a potential threat for it (Heberer 2002, Jørgensen and Halling-Sørensen 2000, Rogers 1996).
Furthermore, the fate of pollutants within the WWTPs is today well studied and WWTP effluents are generally considered as an important source of contamination for a long time, especially in urban areas (Heberer 2002). This implies a better understanding of micropollutants behaviors within wastewater treatment processes.

Generally, wastewater treatment includes pre-treatment, primary and biological treatments. For primary treatments, primary settling (PS) and physico-chemical lamellar settling (PCLS) are commonly used. While PS has been initially and widely employed, PCLS is more and more frequent since this technique operates more compactly and allows a wider flexibility of configuration and use. To our knowledge, some papers exist on one or another technology (Alexander et al. 2012, Carballa et al. 2005, Choubert et al. 2011) but there is no study neither comparing both technologies nor examining the impact of coagulant and flocculant on the pollutant removal at industrial scale.
Concerning biological treatments, they are most of the time designed to achieve a high removal of nitrogenous pollution, and two types of treatments exist. Classical biological units, using suspended growth systems (flocs) such as conventional activated sludge, have already been well documented (Clara et al. 2005, Joss et al. 2005, Katsoyiannis and Samara 2005, Ruel et al. 2010). Moreover, some studies have compared conventional activated sludge (CAS) process with membrane bio-reactor (Bernhard et al. 2006, De Wever et al. 2007, González et al. 2007, Sipma et al. 2010), another suspending growth technology, but only a few of them have compared conventional treatments with biofilm technologies such as biofiltration (Choubert et al. 2011, Joss and Maurer 2006). Biofiltration (BF) is a fixed bed technique consisting in the development of a specific biofilm on a filtration material. Thus, it combines a physical retention of particles and a biological treatment of dissolved molecules by microorganisms. Its compactness (small footprint), modularity (ability to adapt operating parameters to match the wastewater flow) and intensiveness (short hydraulic retention time - HRT) have contributed to the development of this technology since the 80s, particularly in urban areas, where it is the most suitable. Despite these strengths, BF remains very poorly studied regarding efficiency for priority and emerging pollutants.

Within the framework of the OPUR (Observatory of Urban Pollutants, Paris) research program, different studies were carried out on primary and secondary treatments. PCLS and BF were first studied by (Gasperi et al. 2010) for priority pollutants and by (Gilbert et al. 2012) for alkylphenols and polybromodiphenylethers (PBDE). The same methodology was also applied for PS and CAS. More recently, triclosan, triclocarban and parabens were also investigated for both processes (Geara-Matta 2012).
This paper aims at synthetizing and completing all data and knowledge provided by these different studies on two WWTPs treating wastewater from the same catchment (west of Paris). First, each facility performance was assessed to compare PS with PCLS and BF with CAS. Then, overall treatment systems efficiencies were examined. To achieve this goal, the pollutant removals normalized to the nitrogen pollution removals were used, not to correlate micropollutant removal to nitrogen removal, but as a comparison of both treatment systems performances. This normalization is particularly relevant because achieving an efficient nitrogen removal has a huge impact on processes design especially for operational conditions. Thus, this work is innovative because BF is studied and compared to CAS and treatment systems are compared not only by percentage but by efficiency normalized to a parameter widely used by water managers to describe WWTP performances (nitrogen). This methodology enables a relevant comparison of both WWTPs and can provide relevant information since they are principally designed to treat carbonaceous and nitrogenous pollutions.
Therefore, a large panel of molecules including all WFD compounds and some emerging pollutants (n=104) like parabens, biocides (triclosan and triclocarban) or pesticides was monitored. For each compounds, both dissolved and particulate concentrations were measured. Thus, this work provides comparative information about removal performances of processes and explains experimental results by theoretical mechanisms and physico-chemical properties of chemicals. To achieve this goal, the monitoring of conventional parameters like C/N/P was crucial.

METHODS

WWTP description and sampling points

Two WWTPs were studied (Figure 1) upstream (Seine Amont) and downstream (Seine Centre) Paris. Both are run by the Parisian public sanitation service (SIAAP).
The Seine Amont plant receives 600 000 m^3 of wastewater per day. Wastewater is first pre-treated (screening and grit/oil removal), and then settled by PS tanks to remove a large amount of particles. An extended aeration activated sludge unit (biological reactor combined with a secondary settling tank) allows carbon and nitrogen removal. This configuration of activated sludge unit belongs to the most efficient existing one as it operates at very low load (< 0.32 kg BOD_5/(m^3.d), (Gaïd 2008))[1]. The first zone operates in anoxic conditions to remove nitrates and the second zone operates in aerobic conditions and allows the carbon removal and the total nitrification. Finally, the effluent undergoes a tertiary treatment by clariflocculation to complete particle and orthophosphate removal.
The Seine Centre plant receives 240 000 m^3 of wastewater per day and its design consists in a pre-treatment (screening, grit/oil), a PCLS unit (Densadeg®) with coagulant (ferric chloride) and flocculant (anionic polymer) injection, and a

[1] BOD_5: biochemical oxygen demand 5-day test.

three-stage BF unit. The first stage (Biofor® - type filters with biolite as the medium) is designed for carbon removal in aerated conditions, the second one (Biostyr® - type filters with biostyrene as the medium) achieves a total nitrification in aerated conditions and the third one (Biofor® - type filters) consists in a denitrification step in anoxic conditions. This three-stage BF configuration (downstream denitrification) is the most efficient one for nutrients, as shown by (Rocher et al. 2012). Once treatments are achieved, both effluents are discharged into the Seine River.

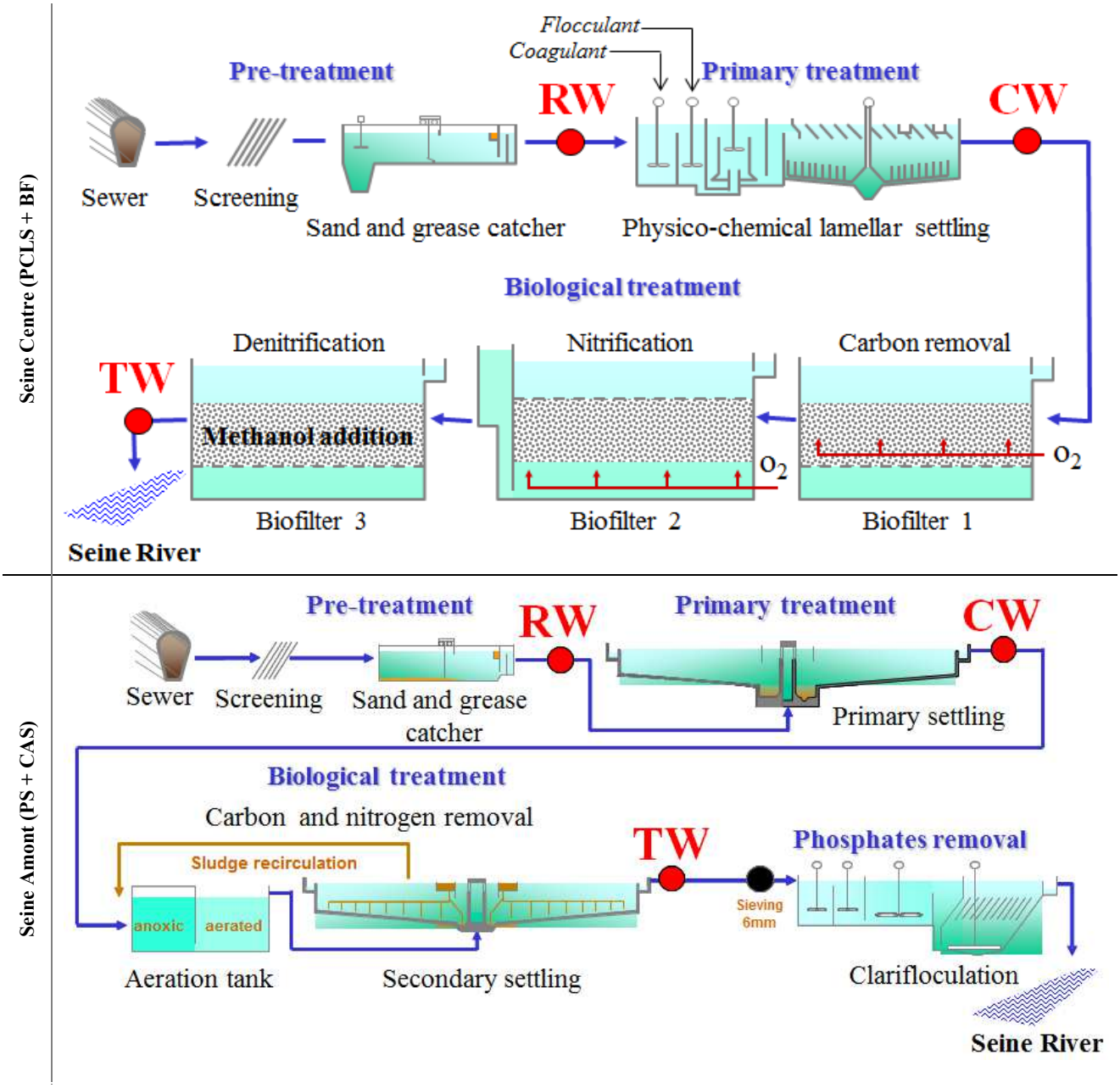

Figure 1. Layouts of the two studied wastewater treatment plants

Three sampling points for both plants were considered: raw water (RW), clarified water (CW) and treated water (TW). Considering the quantity of particles required for analysis (from 0.2 to 2.0 g), large volumes of water were collected (10 L for RW, 30 L for CW and TW) using automatic refrigerated samplers (at 4°C) equipped with glass bottles and Teflon® pipes to avoid any contamination. These samples were 24-h composite samples to be representative. Dissolved fraction was analyzed within 24-h after sampling while particulate fraction was analyzed within 48-h.

Conventional wastewater quality parameters

Table 1 displays concentrations of the main conventional wastewater quality parameters during all sampling campaigns and total removals (R, in %). These parameters include total suspended solids (TSS), chemical oxygen demand (COD),

total Kjeldahl nitrogen (TKN), total nitrogen (TN) and total phosphorous (TP). For each parameter, minimum - maximum and mean (below) values of all campaigns are given. R is calculated between RW and TW.

Table 1. Conventional wastewater quality parameters in RW, CW and TW effluents

	Seine Amont (n=5) PS + CAS				Seine Centre (n=5) PCLS + BF			
	RW	**CW**	**TW**	**R (%)**	**RW**	**CW**	**TW**	**R (%)**
TSS (mg/L)	330.0 - 600.0 426.5	84.0 - 251.3 120.5	4.0 - 14.8 7.8	*95.5 - 99.3* *98.2*	229.0 - 302.6 269.7	24.0 - 65.7 42.1	3.0 - 10.6 6.6	*95.4 - 99.0* *97.6*
COD (mgO_2/L)	564.0 - 1630.0 764.3	249.0 - 365,0 317.8	17.0 - 39.0 25.1	*93.1 - 98.9* *96.7*	426.0 - 546.0 491.3	150.0 - 218.0 175.4	28.3 - 59.0 39.2	*86.2 - 94.8* *92.0*
CODs (mgO_2/L)	-	-	-	-	134.0 - 151.0 139.2	104.3 - 118.7 111.5	23.3 - 32.0 28.1	*76.1 - 84.6* *79.8*
TKN (mgN/L)	56.0 - 78.7 68.6	48.8 - 62.7 57.8	1.2 - 1.9 1.6	*96.6 - 98.5* *97.7*	40.0 - 53.0 47.3	36.0 - 53.0 40.5	1.5 - 8.3 4.8	*79.3 - 97.2* *89.9*
TN (mgN/L)	56.0 - 78.7 68.6	48.8 - 62.7 57.8	14.3 - 23.2 19.8	*58.6 - 81.8* *71.1*	43.0 - 53.9 49.1	38.6 - 45.0 41.9	3.8 - 14.7 11.9	*65.8 - 92.9* *75.8*
TP (mgP/L)	10.0 - 14.2 12.0	7.0 - 9.1 8.0	1.3 - 2.7 2.1	*73.0 - 90.8* *82.5*	5.5 - 7.3 6.3	0.9 - 2.2 1.6	0.2 - 0.4 0.3	*92.7 - 97.3* *95.2*

As shown by mean removals (Table 1), both plants achieve quite similar and high elimination of TSS and COD (> 90%) and to a lesser extent of TN (71-75%) and TP (> 80%). Overall performances obtained with BF process are relevant with annual performances reported by (Rocher et al. 2012). Difference for TP ($TP_{PCLS+BF}$ > 95% vs. TP_{PS+CAS} < 85%) results from precipitation of orthophosphates thanks to coagulant addition in PCLS (Jiang and Graham 1998, Metcalf and Eddy 2003). As regards nitrogen removal, both WWTPs seem as efficient to remove TN in percentage, but actually PS + CAS removes a slightly higher quantity of TN per liter (48.8 mgN/L for PS + CAS vs. 37.2 mgN/L for PCLS + BF) because of a higher raw water concentration (68.6 mgN/L for PS + CAS vs. 49.1 mgN/L for PCLS + BF). In addition, both PS + CAS and PCLS + BF achieve a total nitrification, with high removals of TKN and NH_4^+ (not presented). Finally, both WWTPs are designed to intensively and efficiently remove nitrogen.

Pollutants and analytical procedures

A total of 104 pollutants were monitored. Depending on the substance, two methodologies were deployed. First, a large panel of 81 priority and emerging pollutants (Zgheib et al. 2008) was selected (Table 2) to be screened during three campaigns in 2008 (March, September, December) for PCLS + BF and 2010 (July, November, December) for PS + CAS. Those analyzes were carried out by IPL-Bretagne, a French accredited laboratory (COFRAC - French official accreditation committee). For metals, samples were digested during 2-h using a concentrated mixture of nitric and hydrochloric acids. For organic pollutants, the dissolved compounds were extracted by liquid-liquid extractions (hexane or dichloromethane) for most of them or by solid phase extraction - SPE (polystyrene/divinylbenzene-copolymer cartridges) for pesticides. After lyophilization, particulate matter was extracted by assisted solid extractions (acetonitrile/water for pesticides, hexane/dichloromethane for the remaining organic compounds). Different analytical procedures were used depending on the molecules (Table 2).

Additional analyses were carried out internally on 23 other molecules. Five campaigns were performed for alkylphenols and polybromodiphenylethers (PBDE) in 2010 and three for biocides (triclosan and triclocarbon) and parabens in late 2010 - early 2011. Whatever the period considered and as confirmed by the globally similar removals of conventional wastewater parameters, the operational conditions and performances on both plants were similar.

For alkylphenols and PBDEs, after filtration, the dissolved phases are spiked with alkylphenol deuterated standards (n-OP-d17 and NP1EO-d2) and PBDE quantification standards (BDE 77, BDE 181 and BDE 209-^{13}C). They are then extracted within 24-h by SPE (Oasis HLB® for alkylphenols and C18 Chromabond® for PBDEs). For the particulate fraction, particles are microwave extracted with 20 mL of dichloromethane/methanol (90:10, v/v). Alkylphenols are analyzed by ultra performance liquid chromatography coupled with mass tandem spectrometry (UPLC-MSMS), while PBDEs are analyzed by gas chromatography coupled with mass spectrometer. More details on analytical method are available in (Gilbert et al. 2012). For parabens (6 congeners) and biocides (triclosan and triclocarban – TCS and TCC), acidified dissolved phases are spiked with deuteured internal standards (PrP-d4 - PrP: propylparaben - and TCS-d3), after filtration. Extraction is performed by SPE on Oasis HLB® cartridges. Then, analytes are eluted with 10 mL of methanol and two surrogates are added (MeP-d4 - MeP: methylparaben - and TCC-d4) before analyzing by UPLC-MSMS. Extraction of particular phases is performed by microwave during two successive cycles after spiking by 50 µL of internal standards (TCS-d3 and PrP-d4) from two different solutions. 20 mL of a dichloromethane/methanol (90/10, v/v) mix are added as extraction solvent. After purification, Oasis HLB® cartridges allow to collect two fractions

containing parabens and TCS, or TCC, which are spiked with 50 µL of surrogates (MeP-d4 and TCC-d4) before being analyzed by UPLC-MSMS. More details on analytical method are available in (Geara-Matta 2012). Particulate and dissolved concentrations are measured individually for each sample except for metals and BTEXs/HVOCs analyzed on total fraction.

Table 2. Groups of pollutants and analytical methods

Groups[a]	Total[b]	n[c]	Standards	Methods[d]	Phase[e]
Phenols	2	3	ISO 18857-1	GC-MSMS	P + D
BTEXs	5 (1)	3	NF EN ISO 11423-1	GC-MS	T
Chloroalkanes	1 (1)	3	IPL Internal method	GC-ECD	P + D
Chlorobenzenes	5 (3)	3	EN ISO 6468	GC-MS	P + D
HVOCs	7 (4)	3	NF EN ISO 10301 + 6468	GC-MS	T
PAHs	16 (8)	3	ISO 17993	HPLC-Fluo	P + D
Metals	8 (4)	3	NF EN ISO 11885 + 1483	ICP and AAS	T + D
Organotins	3 (3)	3	NF EN ISO 17353	GC-MS	P + D
PCBs	8	3	NF EN ISO 6468	GC-MSMS	P + D
Phtalates	1 (1)	3	Internal method	GC-MS	P + D
Pesticides	25 (12)	3	NF EN ISO 11369 + IPL Internal method	GC-MS UPLC-MSMS	P + D
Screening	*81 (37)*				
Alkylphenols	6 (2)	5	Internal method (Gilbert et al. 2012)	UPLC-MSMS	P + D
PBDEs	9 (6)	5	Internal method (Gilbert et al. 2012)	GC-MS	P + D
Biocides	2	3	Internal method (Geara-Matta 2012)	UPLC-MSMS	P + D
Parabens	6	3	Internal method (Geara-Matta 2012)	UPLC-MSMS	P + D
Additional analyses	*23 (8)*				

a Groups: BTEXs = benzene, toluene, ethylbenzene and xylenes, HVOCs = halogenated volatile organic compounds, PAHs = polycyclic aromatic hydrocarbons, PBDEs = polybromodiphenylethers, PCBs = polychlorobiphenyls.
b Number of substances listed in the WFD is in bracket.
c Number of campaigns.
d Analytical methods: ICP = inductively coupled plasma, AAS = atomic absorption spectrometry, GC = gas chromatography, GC-ECD = GC with electron capture detector, GC-MS = GC with mass spectrometry, GC-MSMS = GC with tandem mass spectrometry, HPLC-Fluo = high performance liquid chromatography with fluorescent detector, UPLC-MSMS = ultra performance liquid chromatography with tandem mass spectrometry,
e Phase considered with D = dissolved, P = particulate, T = total/bulk sample.

RESULTS AND DISCUSSION

Comparison of primary treatments performances

The removals (in %) of priority and emerging pollutants, grouped by families (vertical lines), by PS and PCLS are illustrated in
Figure 2 for each campaign (n=3-5, Table 2). Out of the 104 molecules monitored, 68 were detected in raw water. Globally, the pollutant pattern found and the orders of magnitude are quite comparable with those reported in literature (Clarke et al. 2010, Deblonde et al. 2011, Fatone et al. 2011, Gasperi et al. 2008, Karvelas et al. 2003, Komori et al. 2006). As expected, high levels of metals (12-354 µg/L), DEHP (13-66 µg/L), BTEXs/HVOCs (0.65-7.5 µg/L), chloroalkanes (38-59 µg/L) and alkylphenols (2.71-3.57 µg/L) were found (Supplementary data - Table 1). Similarly, parabens (1.79-7.38 µg/L) and TCS (2.82-6.58 µg/L) were found at high levels, preferentially in dissolved fraction (> 94%) for parabens. This is interesting as their occurrence in French wastewater is still not well documented. Finally, a quite similar quality was observed in influent of both WWTPs for the majority of compounds, resulting in a similar micropollutant pattern.

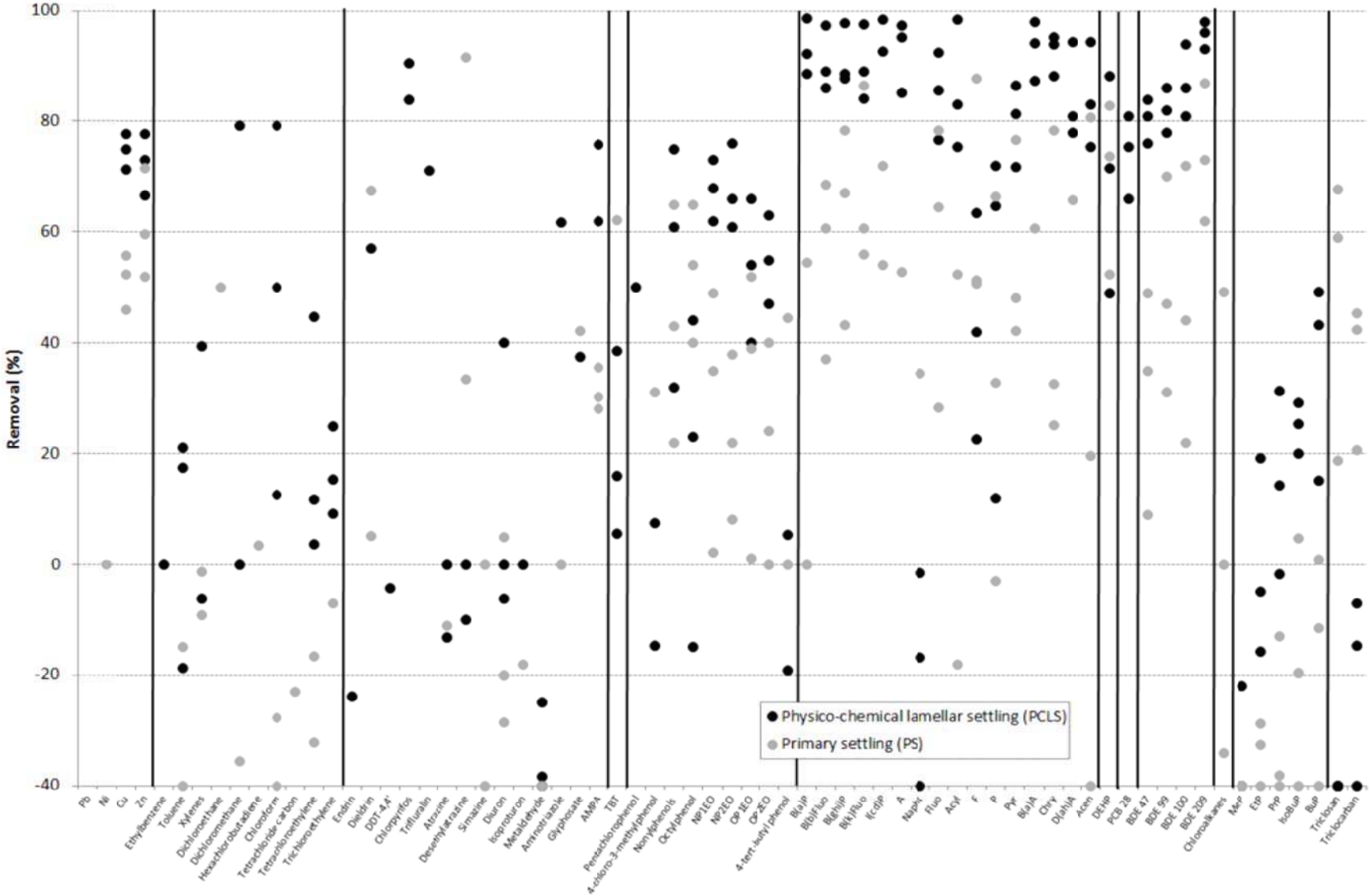

Figure 2. Removal of priority and emerging pollutants by primary treatments

Figure 2 displays the high variation of removal for the majority of compounds. Except for metals or some PAHs and PBDEs exhibiting high and less variable removals, most of pollutants show removals varying from 20 to 60%. These variations appear weaker in the PCLS unit than in PS unit, maybe because of chemicals added which may have a stabilization effect as particles removal is more efficient and homogenous in the unit. Despite the high variations, three groups are distinguishable regarding their removals. Globally, hydrophobic pollutants (log K_{ow} > 4) like PCB-28, DEHP, PBDEs or high molecular weight PAHs, are well or very well removed (> 70%) while hydrophilic ones (log K_{ow} < 3-4) are not or just slightly removed (pesticides, parabens, BTEXs/HVOCs). Alkylphenols, metals (zinc, copper), biocides and tributyltin are moderately removed (20-70%). These removal results are in accordance with other studies (Bergé et al. 2012, Choubert et al. 2011, Gasperi et al. 2010). Moreover,
Figure 2 displays higher removals (10-30%) with PCLS than PS, reflecting the impact of coagulation/flocculation on removal of micropollutants. This can mainly be explained by an improvement of TSS removal. Actually, the difference in hydrophobic compounds and metals removal rates (about 20%) is directly correlated to the better settling of particles in PCLS (Table 1). Improvement of particulate pollutant removal by coagulation/flocculation has already been highlighted in the literature (Alexander et al. 2012, Bratby 2006, Duan and Gregory 2003, Gasperi et al. 2012).

For the dissolved fraction, there is no clear trend displaying a benefit from chemicals in PCLS especially because of the high variations of removal, even if coagulant and flocculant have an impact on CODs (10-20%) and PO_4^{3-} (10-15%) removal (Table 1). The removal of dissolved fraction of organic pollutants induced by coagulation/flocculation has already been displayed (Bratby 2006, Vigneswaran et al. 2009). Similarly, some soluble compounds are slightly better removed (10-20%) in presence of coagulant and flocculant like alkylphenols, PBDEs or parabens (
Figure 2). This can be explained by adsorption on flocs and/or impact of coagulation/flocculation on colloids which can be a sorption site for some pollutants by hydrophobic interactions or adsorption (Bratby 2006, Elimelech et al. 1995, Vigneswaran et al. 2009). Moreover, a precipitation mechanism can occur, in which coagulant neutralizes anionic sites of organic molecules, changing radically their solubility (Duan and Gregory 2003). The colloidal fraction elimination was mentioned for endocrine disrupting compounds (Zhou et al. 2006) and PBDE (Song et al. 2006), or proved for metals (Li et al. 2002). For the treatment of combined sewer overflows by ballasted clariflocculation, (Gasperi et al. 2012) also observed a more or less marked removal of dissolved fraction. Similarly, authors reported high variations from pollutant to pollutant and for a given pollutant. However, the impact of coagulant and flocculant is higher by far on the particulate phase than on the dissolved phase in terms of micropollutants removal.

Biological treatments

Figure 3 illustrates the removals for each pollutant by CAS and BF (n=3-5) treatments, ranked by families (vertical lines).

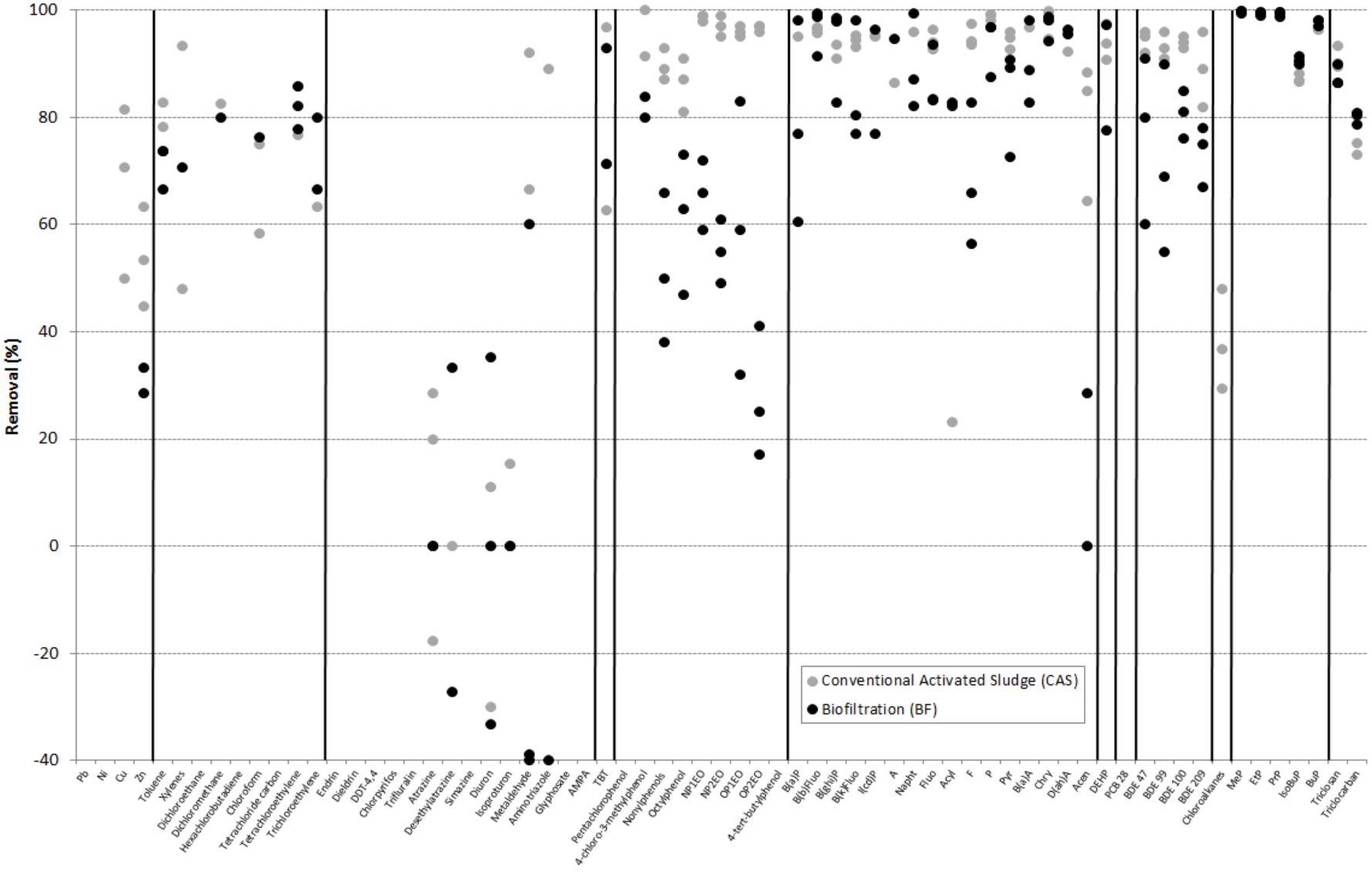

Figure 3. Removal of priority and emerging pollutants by biological treatments

First of all,
Figure 3 displays a much weaker variation of removal for most pollutants than for primary treatments, except for pesticides. Removals are quite stable for both units with a maximal variation of 20% most of the time. However, these variations seem slightly weaker with CAS than with BF. This could be due to the lower HRT in BF units (20-30 h for CAS vs. 45-60 min for BF), resulting in influent peak loads being less averaged over time, according to the lower reactor volume. All compounds are eliminated from moderately to efficiently (20-80%, or > 80%), except pesticides which are not removed by any units. For pesticides, this is in good accordance with the study of (Ruel et al. 2012). Results for biocides and parabens are particularly interesting as variations are very low and removals very high for both units (> 70-80% for biocides and > 90% for parabens). This is a quite important observation regarding the lack of literature data about their occurrence and fate in WWTPs. Nevertheless, high removals (> 90%) have also been reported for parabens (Andersen et al. 2007), mainly during biological treatments (Eriksson et al. 2009), and for biocides with BF or CAS (Heidler and Halden 2007, Sabaliunas et al. 2003).

Both biological systems (CAS and BF) give comparable results despite different inlet concentrations, what is consistent with results of (Choubert et al. 2011). They obtained removals in the same range for 70% of the 125 molecules they searched (pesticides, pharmaceuticals, PAHs, phtalates, alkylphenols, HVOCs, metals, etc.) with 2 BF and 5 low load CAS WWTPs. The differences between BF and CAS were only observed for some pharmaceuticals, explained by better biodegradation. In our case, it is particularly true for BTEXs/HVOCs, PAHs, DEHP, biocides, parabens and tributyltin. Globally, moderate or high removals for PAHs and PBDEs were also reported in the literature (Clarke et al. 2010, Fatone et al. 2011, Manoli and Samara 2008, Rayne and Ikonomou 2005, Song et al. 2006). The very high elimination (> 80%) of hydrophobic compounds (log K_{ow} > 4) is consistent with high TSS removal in both systems (85% with CAS; 94% with BF), and it highlights the removal mechanism of sorption of dissolved fraction on particles trapped in biological reactor. Volatile molecules like BTEXs and HVOCs have also high removals (> 70%) in both systems which are intensively aerated allowing transfers to atmosphere by air stripping. However, tetrachloroethylene is always detected in BF effluent contrary to CAS but it can be explained by a significantly higher inlet concentration. This is in accordance with the domestic origin of wastewater in the BF WWTP which classically contains high levels due to dry cleaning equipment discharges (Lohman 2002). Biodegradation can be highlighted by the decrease of dissolved concentration observed in both processes for some biodegradable compounds (DEHP, some PAHs, PBDEs and alkylphenols). Decrease of dissolved concentration for alkylphenols, like nonylphenols (NP), octylphenol (OP) or nonylphenol monoethoxylate (NP1EO), is clearly higher in CAS. In fact, other studies displayed the biodegradation of these compounds: PAHs (McNally et al. 1998), alkylphenols (Bertanza et al. 2011, Clara et al. 2007, Ying et al. 2002) PBDEs (Langford et al. 2007) and DEHP (Bergé et al. 2013, Fauser et al. 2003). Finally, the three main removal mechanisms (biodegradation, sorption and volatilization) identified in the literature (Byrns 2001, Cirja 2008, Mozo et al. 2012) for biological treatments are highlighted by this study.

Even if globally both biological units exhibit comparable efficiencies, some compounds are slightly better removed by CAS like Zn, 4-chloro-3-methylphenol, alkylphenols and PBDEs (to a lesser extent). Alkylphenols better removal by CAS, confirmed by a higher dissolved concentration decrease, could be explained by biodegradation and sorption mechanisms. In fact, a higher biodegradation could be expected in CAS, regarding its higher HRT, as well as a higher sorption on activated sludge flocs than on biofilm due to the physico-chemical properties of biomass (floc size and morphology, specific surface, etc.). Organic pollutants like DEHP, alkylphenols or PBDEs may be potentially more sorbed on activated sludge similarly to metals for which it was observed (Tian et al. 2006, Wang et al. 2010). Specific studies comparing sorption capacity of biofilms and activated sludge flocs should be held to validate this hypothesis. However, (Clara et al. 2007) showed that biodegradation is the main removal pathway for alkylphenols in CAS WWTPs, representing more than 85% of the removal while sorption onto sludge represents only 15% of it. This tends to validate the hypothesis of a biodegradation enhancement by CAS compared to BF.

Comparison of WWTP performances

According to the biological treatment chosen, requirements for clarified effluents vary. CAS needs the presence of biodegradable carbon in influent for denitrification whereas BF requires a quite low level of TSS to avoid operational problems like fouling. That is why the more intense unit regarding particles removal (PCLS) precedes BF as primary treatment while the less intense unit (PS) precedes CAS. Given this, the comparison of treatment systems (primary + biological treatments) gives different information about efficiency and appears more relevant than simple comparison of units. The global performances can be evaluated conventionally (in %), but in this study a more relevant method is also used. Actually, the quantity of pollutant removed per unit of nitrogen removed was calculated (Figure 4 and Figure 5). Moreover, the choice of nitrogen as normalization parameter rather than COD or TSS is logical since the treatment of nitrogen in water implies important modifications on processes (biomass nature and diversity, hydraulic and sludge retention times) in comparison to WWTPs treating carbonaceous and particulate pollution. This parameter represents then a good indicator of the global operation of current WWTPs. Thus, quantity of each micropollutant removed

between RW and TW was calculated and divided by the quantity of nitrogen removed in the process. About 48.8 mgN/L of TN are removed by PS + CAS while PCLS + BF allows about 37.2 mgN/L of TN removal (Table 1).

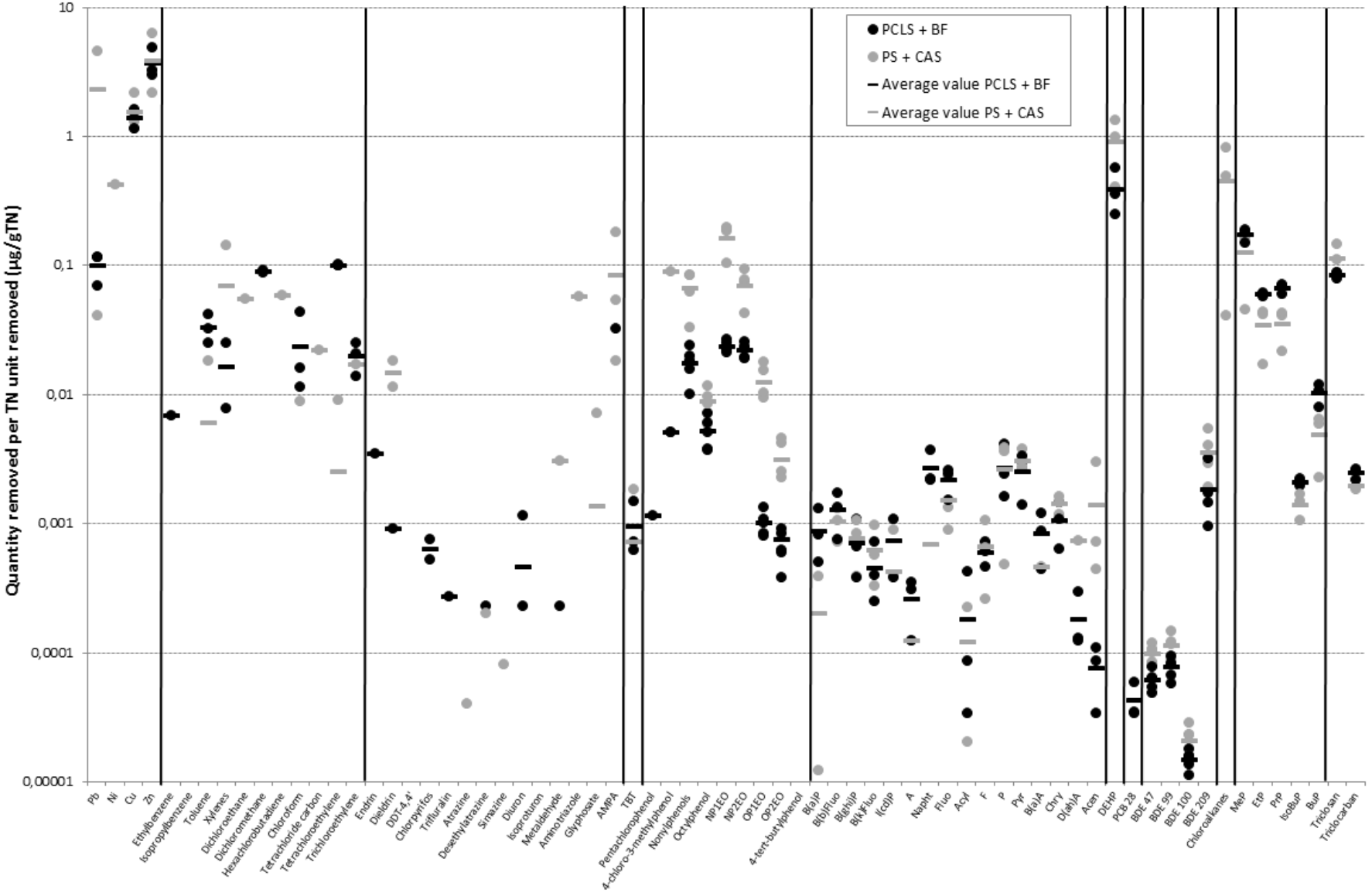

Figure 4. Micropollutant removal normalized to nitrogen removal in both WWTPs

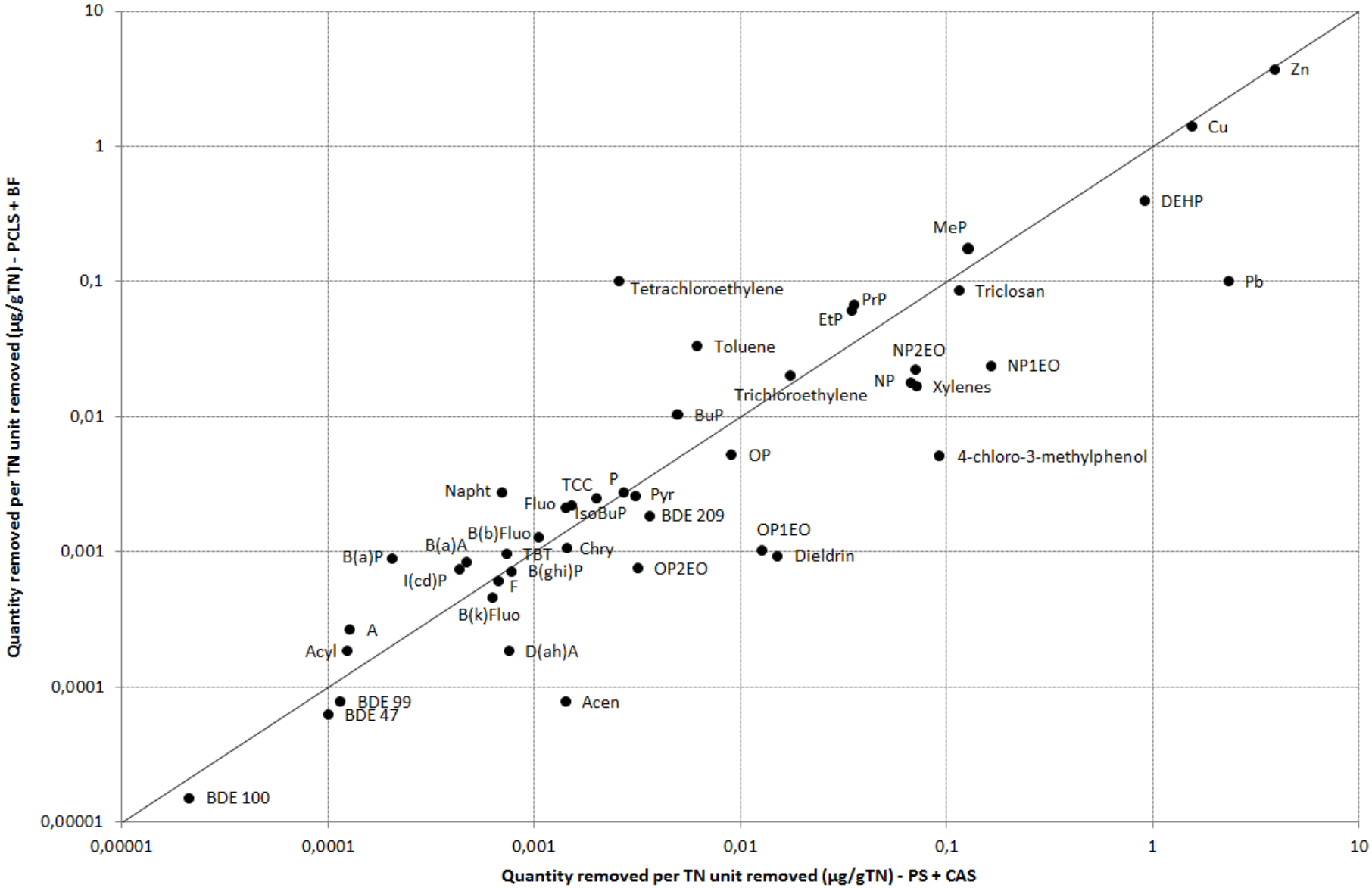

Figure 5. Comparison of both WWTPs efficiencies for micropollutants regarding removal normalized to nitrogen removal

According to
Figure 4, variations of results are quite weak in both WWTPs except for pesticides which are never removed. Even so, some pollutant removals exhibit higher variations, which may originate from high RW concentration variations (Pb and xylenes) or from inherent variations in removal mechanisms (acenaphtene - Acen and acenaphtylene - Acyl). Finally, variations of removal seem slightly higher for PS + CAS than PCLS + BF, resulting from higher variations in PS than PCLS (stabilization effect of chemicals). This representation allows observing that when 1 g of TN is eliminated, a removal of 0.5-1 µg of DEHP, 0.02-0.07 µg of NP, 0.07-0.12 µg of TCS, 0.1-0.3 µg of MeP or 3.7-3.9 µg of Zn could be expected in such WWTPs. To our knowledge, this kind of information is not available yet but can be used to predict pollutant removal in WWTPs.

Globally, both treatment systems have similar performances regarding removals of BTEXs/HVOCs, PAHs, metals (Cu and Zn), pesticides (poorly removed), biocides and TBT. Thus, comparable efficiencies are obtained for the majority of compounds despite a higher compactness and a much lower HRT for PCLS + BF. This is confirmed by Figure 5 where almost all mean removals are distributed along the y = x straight line (same µg of pollutant removed for 1 g of TN removed). This confirms previous observations (Göbel et al. 2007, Joss et al. 2004) reporting comparable efficiencies (in %) between CAS and fixed bed WWTPs for some well removed micropollutants. Moreover, both WWTPs have a very high and comparable efficiency for TSS, resulting in a high and comparable removal of particulate fraction of micropollutants, which is the main pathway for the more hydrophobic compounds.

Some pollutants are better removed by one or another system. Overall, PS + CAS seems allowing a better removal per nitrogen removed for alkylphenols, DEHP, PBDEs, 4-chloro-3-methylphenol and Pb, while tetrachloroethylene and parabens (slight difference) seem to be better eliminated by PCLS + BF. Differences for tetrachloroethylene and Pb can be explained by their significant difference of RW concentration in both WWTPs (Supplementary data - Table 1). For parabens, a slightly better performance of PCLS + BF is observed since a slightly higher quantity of TN (Table 1) was removed within this WWTP during the campaigns performed. In contrary, the better removal of alkylphenols, PBDEs, DEHP and 4-chloro-3-methylphenol by PS + CAS system comes from removal mechanisms. As efficiencies over TSS in both WWTPs are equivalent (> 95%, Table 1), the difference tends to highlight the better removal of dissolved fraction (dissolved + colloidal) of pollutants in the CAS unit, as previously stated.

Actually, dissolved pollutants can be removed by different mechanisms, like sorption on sludge or biodegradation. As exposed in the literature, and more especially for CAS, biodegradation can be affected by different parameters like HRT or concentration, diversity and activity of biomass (McAdam et al. 2010a). Biomass characteristics are strongly affected by conditions required for the growth of nitrifying biomass (HRT and SRT), leading to a better dissolved pollutant removal than other biological conditions (Clara et al. 2005, McAdam et al. 2010a). Nitrification is then a crucial step for dissolved micropollutants removal and total nitrification allows higher removal than partial nitrification as it was observed in literature (Margot et al. 2011). In the total nitrification configuration, biodegradation is enhanced (Clara et al. 2005, Margot et al. 2011) especially through stimulation of micropollutant cometabolism (Carballa et al. 2011, Fernandez-Fontaina et al. 2012). Biomass is affected by SRT which increases the adaptation and diversity of microorganism (Cirja et al. 2008). (Clara et al. 2005) reported a critical value of 10-15 days for the removal of the majority of biodegradable compounds by CAS, which is in the same range as SRT required for nitrification (at least 15-18 days, (Carballa et al. 2011)). Furthermore, HRT has a proven impact on removal of biodegradable compounds as it drives the reaction time (Fernandez-Fontaina et al. 2012, Vieno et al. 2007).

In our case, both PS + CAS and PCLS + BF globally achieve comparable removal of classical wastewater quality parameters such as TSS and TN (Table 1). In particular, they both operate in the most favorable configuration, total nitrification. Nevertheless, HRT in PS + CAS is more than 20 times higher than in PCLS + BF, which theoretically and for the above mentioned raisons would lead to highly different results. Contrariwise, results for biodegradable compounds previously cited (alkylphenols, PBDEs, DEHP and 4-chloro-3-methylphenol) are not hugely different between both WWTPs, even if the CAS WWTP is slightly more efficient, displaying the existence of a process balancing the very short HRT. The difference of biomass structure, which could allow a higher intensiveness of biodegradation in the case of a biofilm, may be this process, but specific measurements are required to demonstrate this assumption.

The slight difference of efficiency in favor of the the CAS WWTP can then be logically explained by the HRT, as observed in the literature for pharmaceuticals (Carballa et al. 2011, Fernandez-Fontaina et al. 2012, Joss et al. 2008) and alkylphenols (McAdam et al. 2010b) within different CAS WWTPs, or to a difference of sorption capacity. (Mahendran et al. 2012) have compared activated sludge flocs and biofilm from a unique water treatment biological reactor and they have shown that flocs were more hydrophobic and negatively charged, in addition to their higher quantity of extracellular polymeric substances (EPS). EPS plays a crucial role in the removal of micropollutants as they represent the main sorption pathway thanks to binding sites they contain (Sheng et al. 2010). These observations tend to indicate a

probable higher propensity to sorb on activated sludge flocs than on biofiltration biofilm for micropollutants. This could lead to a higher biodegradation as a part of micropollutants are degraded when sorbed to the biomass, depending on the compounds (Pomiès et al. 2013).

CONCLUSIONS

This study has investigated the differences in removal of priority and emerging pollutants between two main WWTP treatment systems. PS + CAS represents the most common system, well studied and known whereas PCLS + BF is still widely unknown concerning micropollutants despite its practical increasing interest (compactness, modularity and intensiveness).

As concern the two primary treatments, coagulation/flocculation offers a real gain in terms of micropollutants removal. This gain occurs mainly on particulate pollutants by the way of TSS removal, even if a slight improvement seems to be possible for some groups of soluble pollutants, removed with the colloids. Despite its existence, this effect is not obvious and clear because of the high variations of results. Jar test and laboratory test are maybe requested to really demonstrate the impact of coagulant and flocculant. The partitioning of pollutants in the dissolved phase, between colloidal and soluble fraction, has also to be better studied to characterize the precise effect of coagulation/flocculation on micropollutants. BF appears to be able to remove most of micropollutants as efficiently as CAS in percentage. Yet, some pollutants are slightly better removed by CAS (alkylphenols, metals, some PAHs, 4-chloro-3-methylphenol and PBDEs) due to better biodegradation and/or sorption.

Considering the treatment systems, both configurations seem as efficient but removals between primary and secondary treatments vary. This tendency is even more obvious when removals are represented per nitrogen removed; both systems are globally comparable at equivalent nitrogen removal. Some biodegradable compounds are rather slightly better removed by PS + CAS thanks to higher HRT and flocs characteristics. This normalization could appear relevant in the future since WWTPs are generally designed to treat nitrogen, so the knowing of efficiency for this parameter could allow estimating efficiency for micropollutants. Removal depends on influent concentration as all molecules with comparable concentrations in raw water are removed comparably in quantity, but also on nitrification rate. The development of such approaches could be useful for WWTP managers.

Finally, in the water discharged, most of compounds are not detected or just promptly. In particular, many compounds detected in raw water are never detected in treated water, showing the positive effect of wastewater treatments on many micropollutants. Despite that, some environmentally harmful species are still present at a µg/L level, like metals, pesticides, DEHP or chloroalkanes, because of their high influent concentration or the weakness of treatments on them (pesticides). Although the majority of compounds are below EQS in discharges, TBT, BDE 209 and chloroalkanes are found at concentrations significantly higher than their EQS (factor 5-10) while diuron, chloroform, NP and OP are very close from them. This issue incites to reinforce the idea of the existing installations improvement and/or addition of a tertiary treatment to complete their elimination.

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